



AIMAC collaboration



Research and Development for Accuracy Improvement of Neutron Nuclear Data on Minor Actinides

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Present study includes the result of “Research and Development for accuracy improvement of neutron nuclear data on minor actinides” entrusted to the Japan Atomic Energy Agency by the Ministry of Education, Culture, Sports, Science and Technology of Japan (MEXT).

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Contents

- Needs and Recent Situation of MA data
- Aim, Actions, Approach of the AIMAC
- Highlights of the AIMAC

Details

S033, S055, S111, S114, R130, S185,
R300, S301

- Conclusions and Future Perspective

Needs of accurate nuclear data of MA for nuclear transmutation technology

Example:

Uncertainty analysis on
JAEA-ADS Criticality

dk with

Covariance of JENDL-4.0

$$\frac{dk}{k} = 1.0\%$$

$$1 + k + k^2 + \dots = \frac{1}{1 - k}$$

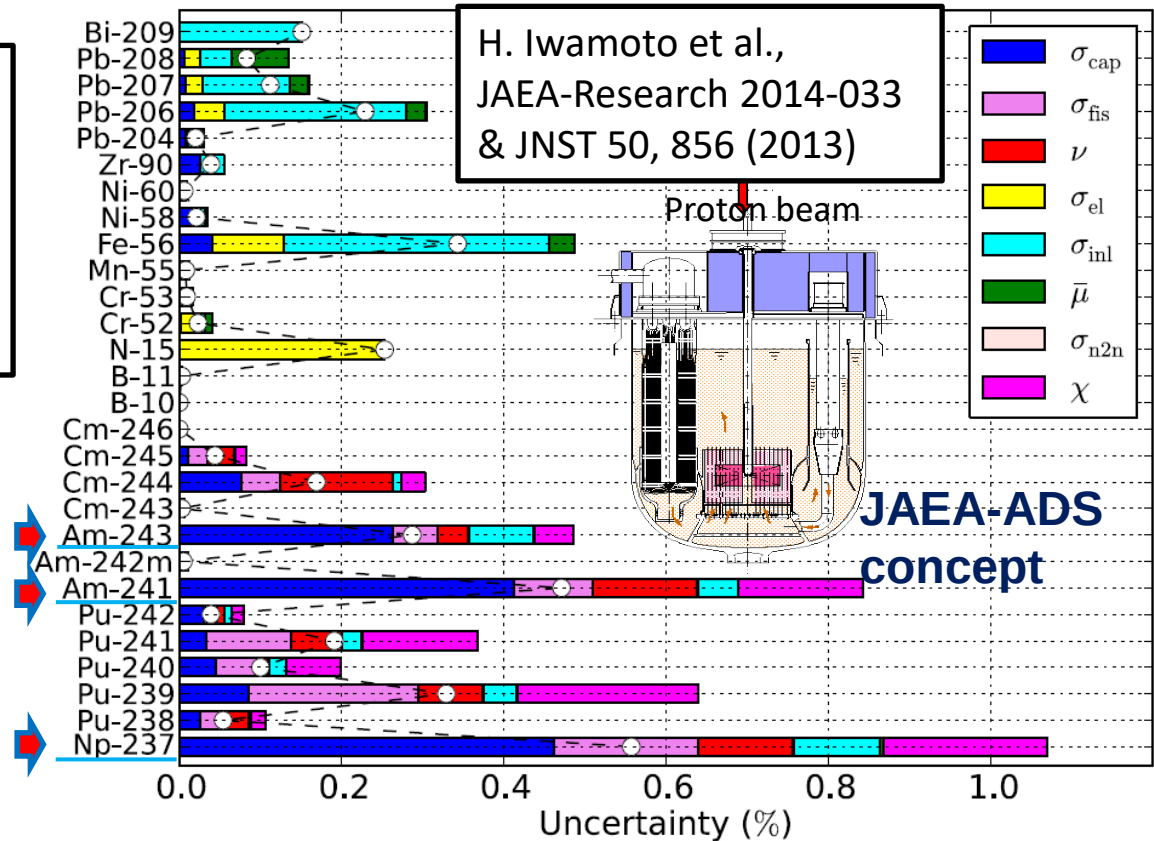
$$k = 0.95 \quad 20$$

$$k = 0.96 \quad 25$$

$$k = 0.97 \quad 33$$

$$k = 0.98 \quad 50$$

$$k = 0.99 \quad 100$$



Accuracy improvement of
Capture of Am-241, Am-243, Np-237
Significant impact!

Recent situation on MA capture

Evaluation	^{241}Am thermal neutron capture cross section
JEFF-3.1.2 $\rightarrow$ JEFF-3.2	647 $\rightarrow$ 748 b
ENDF/B IV $\rightarrow$ VII.0 $\rightarrow$ VII.1	582 $\rightarrow$ 619 $\rightarrow$ 684 b
JENDL-2 $\rightarrow$ 3.3 $\rightarrow$ 4.0	600 $\rightarrow$ 640 $\rightarrow$ 684 b

Important progress
presented at ND2013 on
 ^{241}Am capture.

Good Energy Dependence
Bias due to Westcott factor
Impact of Transmission data
Cold neutron beam data

▪ Still problem on **Absolute**

Int. Nuclear Data Project
OECD/NEA/WPEC/SG-41
(**INDA**) Dr. G. Žerovnik R119

Japanese Nuclear Data Project
AIMAC ^{237}Np , ^{241}Am , ^{243}Am
S033, S055, S111, S114, R130,
S185, R300, S301

The others also n_TOF ...

Aim and Actions of AIMAC

Japanese national nuclear data project

October 2013 – March 2017

Aim of AIMAC:

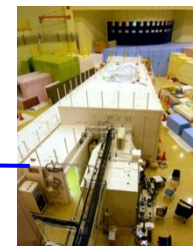
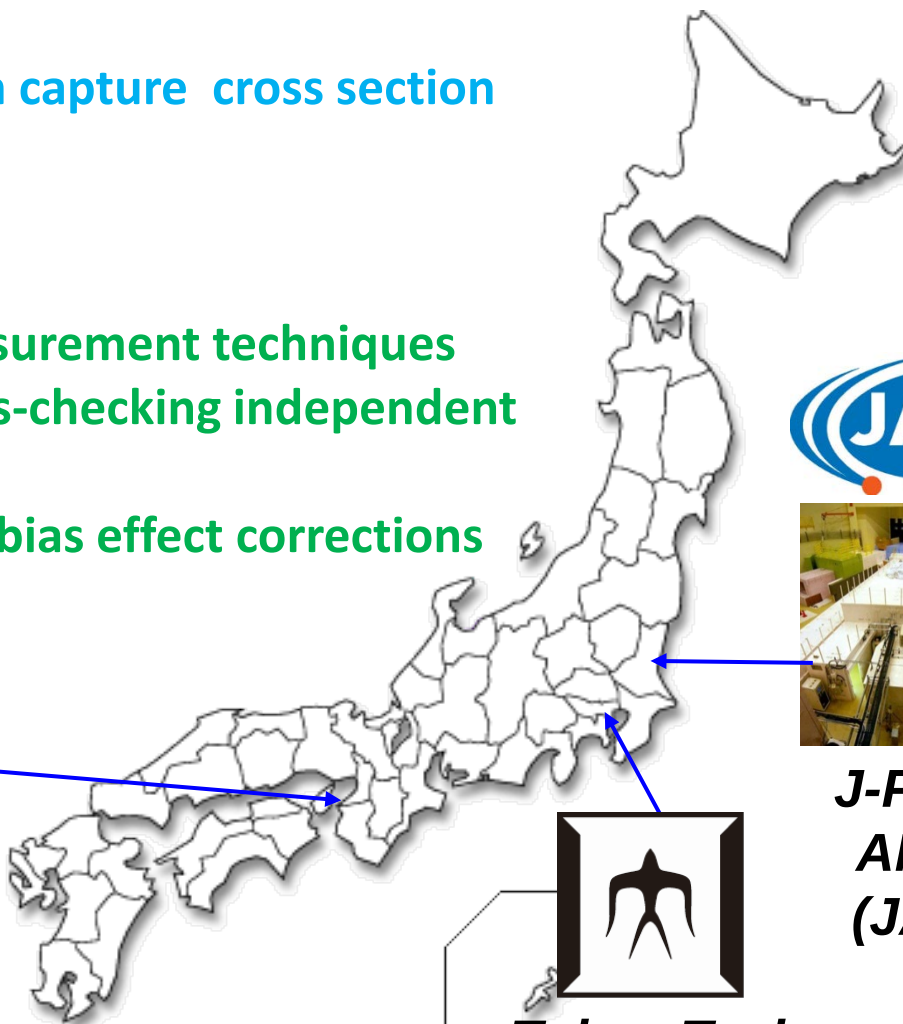
Improve accuracy two times on capture cross section
of Am-241, Am-243, Np-237

Actions of AIMAC:

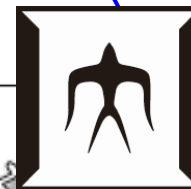
- Develop plural precise measurement techniques
- Identify bias effects by cross-checking independent measurements
- Systematic evaluation with bias effect corrections



KURRI-LINAC



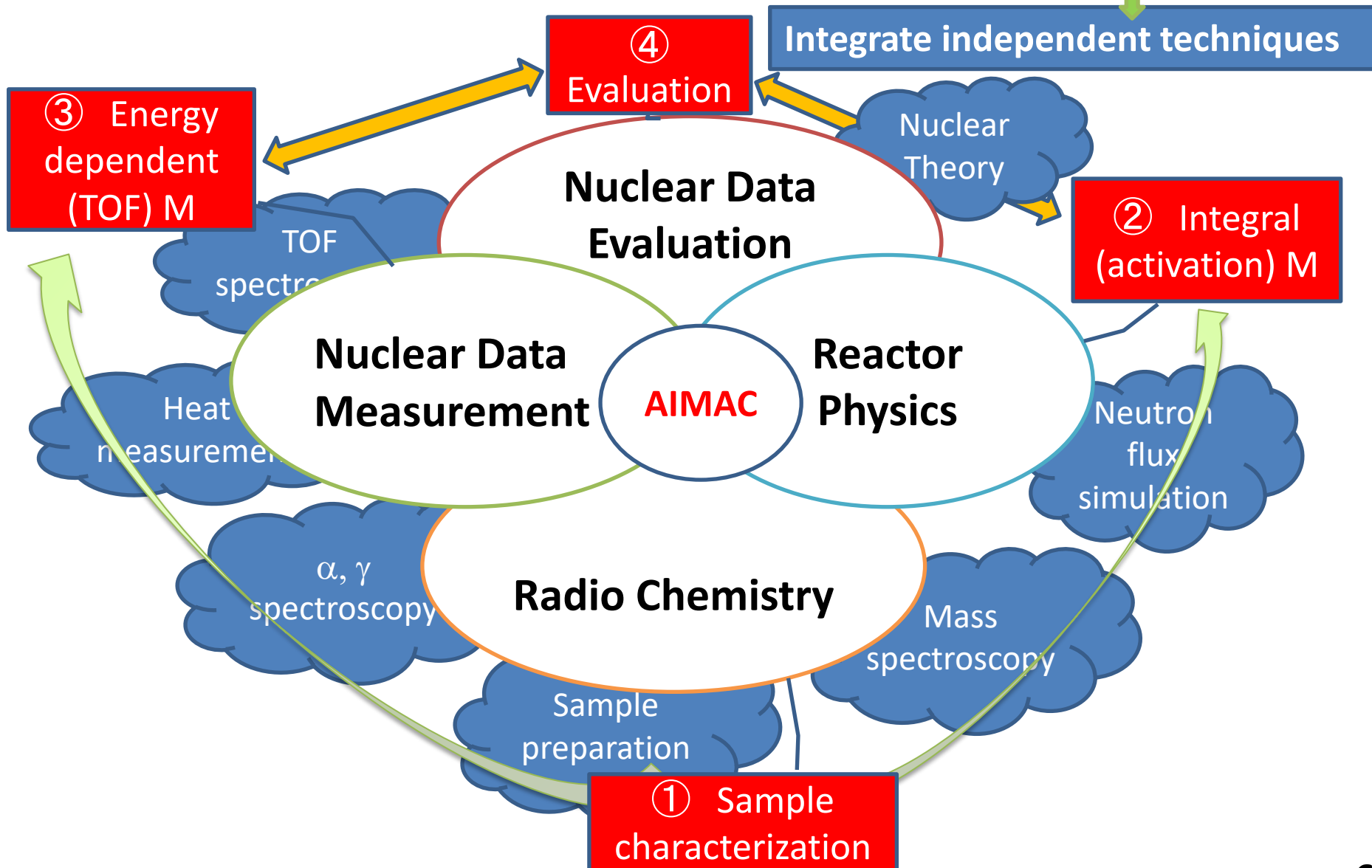
**J-PARC-
ANNRI
(JAEA)**



Tokyo Tech

AIMAC approach

Researchers from 4 different fields tightly collaborate



Highlights of the AIMAC

- ① Precise characterization of sample
- ② Activation measurement
- ③ TOF measurement
- ④ Evaluation and Bias effect Correction

① Precise characterization of sample

- For isotopic purity characterization -

The same materials with TOF samples were prepared for destructive analyses: Mass (TIMS) and α -ray spectroscopy



**Sample
for
analyses**

The Same
Sample
material



**TOF
sealed
sample**

Info in specification sheets:

Am-241 sample (130 kBq/mL)

Isotopic purity: Am-241 : 99.9%

Chemical purity: 99.9% (Pu-239 : 0.09%, Np-237 : 0.01%)

Am-243 sample (10 kBq/mL)

Isotopic purity: Am-243 : 97.3%, Am-241 : 2.7%

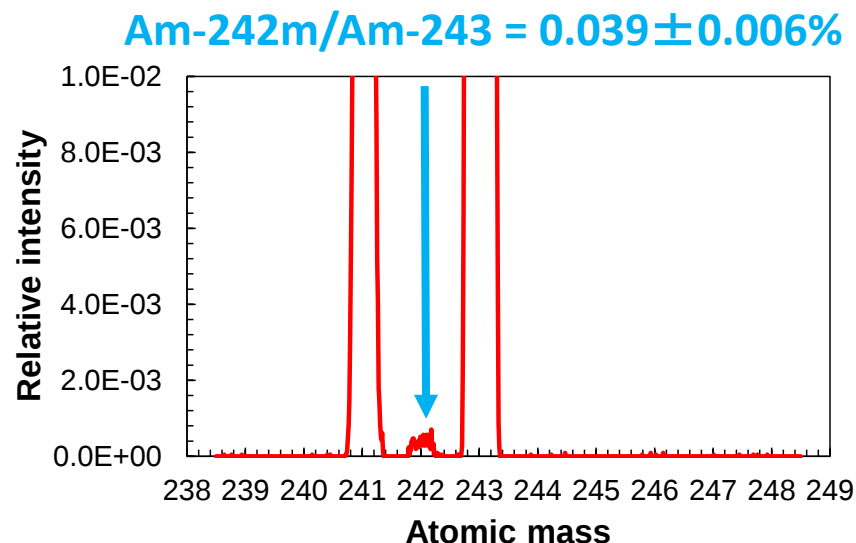
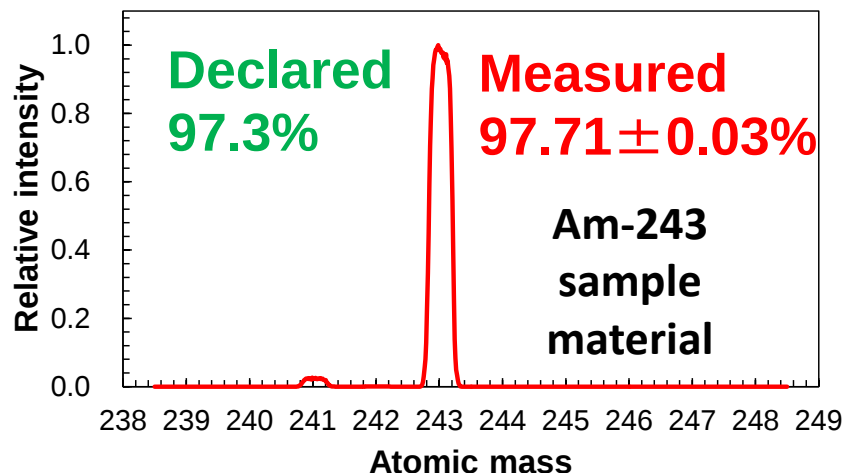
Chemical purity: 99.58%

(Fe, Ca, Na : 0.07%, B : 0.09%, Gd : 0.03% etc)

**No uncertainty
information**

① Precise characterization of sample

Thermal ionization mass spectroscopy (TIMS)



	Am-241	Am-243
Run-1	$2.29 \pm 0.03\%$	$97.71 \pm 0.03\%$
Run-2	$2.28 \pm 0.03\%$	$97.72 \pm 0.03\%$
Run-3	$2.29 \pm 0.05\%$	$97.71 \pm 0.05\%$

Details S301 by
Dr. Y. Shibahara



0.03% precision !

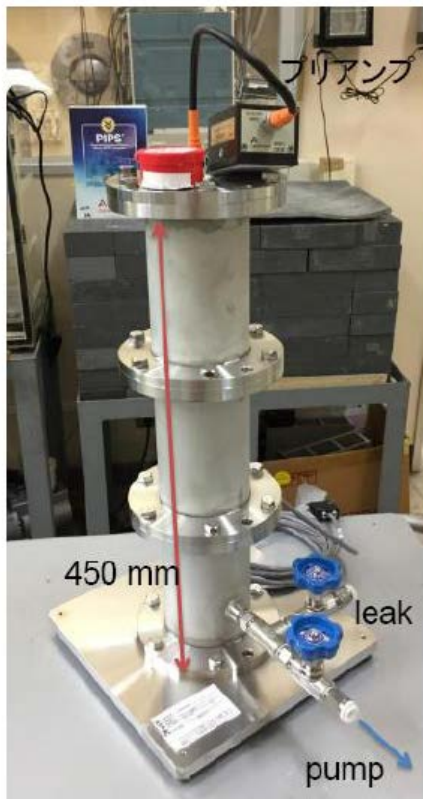
Non-declared isotope
was identified.

Note: Pu contamination
under investigation

Ref: Y. Shibahara, J. Radioanal.
Nucl. Chem. 307, 2281 (2016)

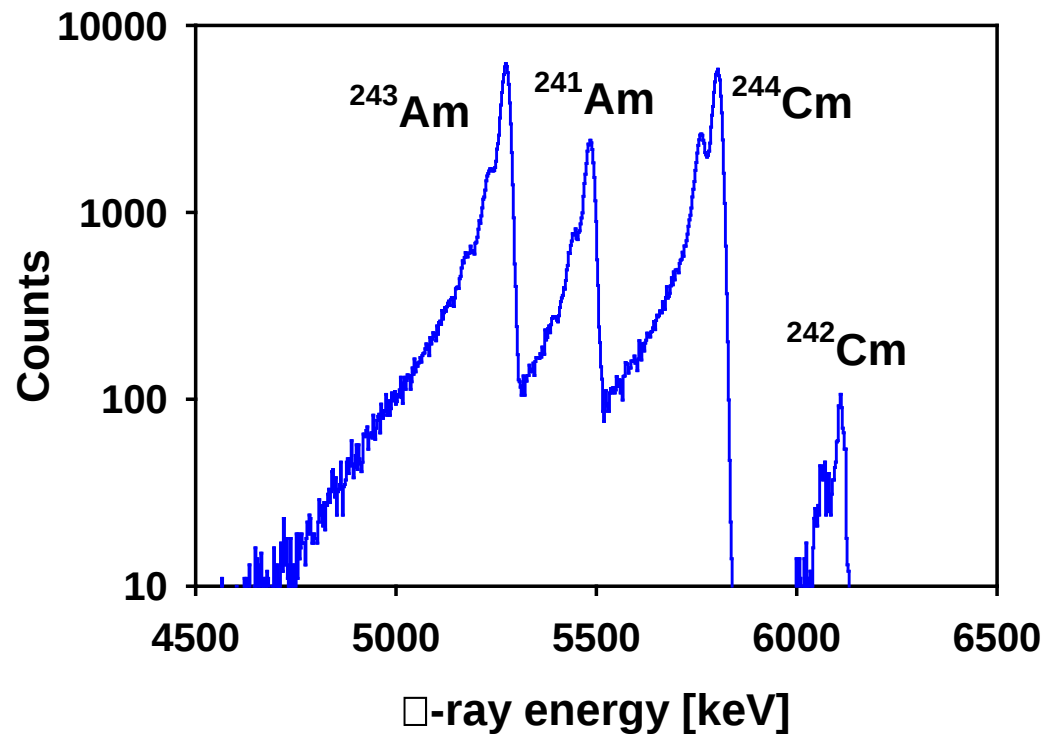
① Precise characterization of sample

α -ray spectroscopy



Setup for α -ray spectroscopy

α -ray from **Am-243** sample material



Cm-242&244 quantified



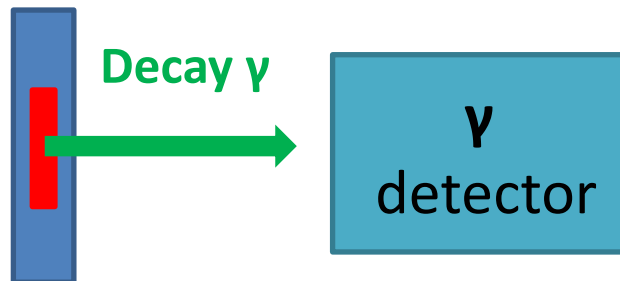
Feedback to Micro Calorimetry

① Precise characterization of sample

No uncertainty information for sealed sample amounts
(decay γ -ray rate is given by 10% uncertainty)

Two non-destructive methods developed

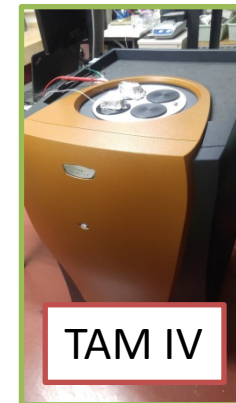
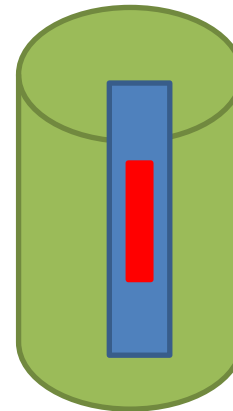
① Determination of decay γ -ray intensity



Precise determination of γ -detector's peak efficiency
Fine interpolation method was developed by utilizing Monte Carlo

Precise determination of γ -ray emission probability

② Utilization of Micro Calorimetry



Independent amounts
Cross-check

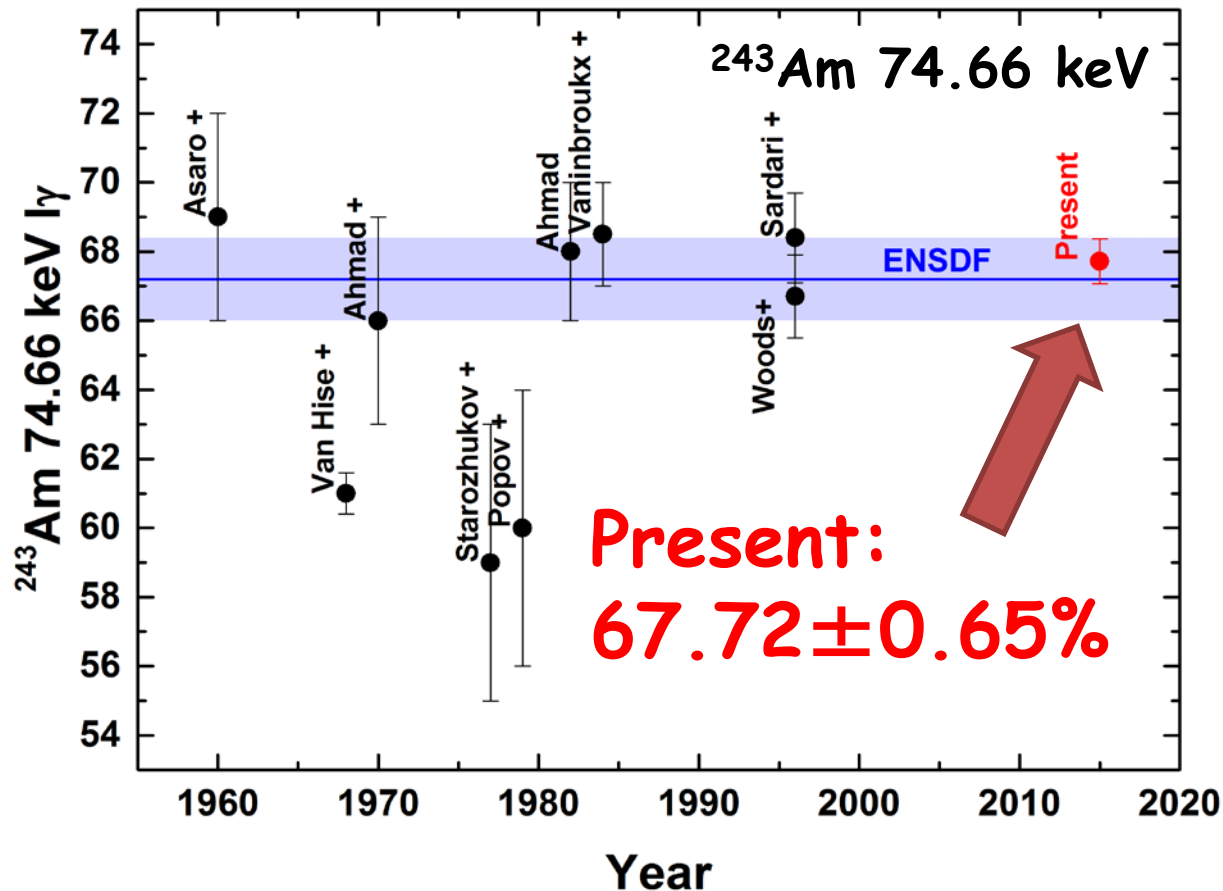
① Precise characterization of sample

- Precise determination of γ -ray emission probability -

Precise determination of γ -detector's peak efficiency

Fine interpolation method was developed by utilizing Monte Carlo

Dr. K. Terada, *J. Nucl. Sci. Technol.* **53** (2016)



① Precise characterization of sample

Comparison of two independent measurements

Declared Activity	Am-241 480 MBq	Am-241 950 MBq	Am-243 60 MBq	Am-243 120 MBq	Am-243 240 MBq
γ -ray Spectroscopy	511 ± 10	962 ± 19	66.7 ± 1.3	155 ± 3	286 ± 6
Micro Calorimetry	510.7 ± 0.5	957.4 ± 0.5	$67.3 \pm 0.3^*$	$155.8 \pm 0.3^*$	$281.8 \pm 0.3^*$

2% uncertainty

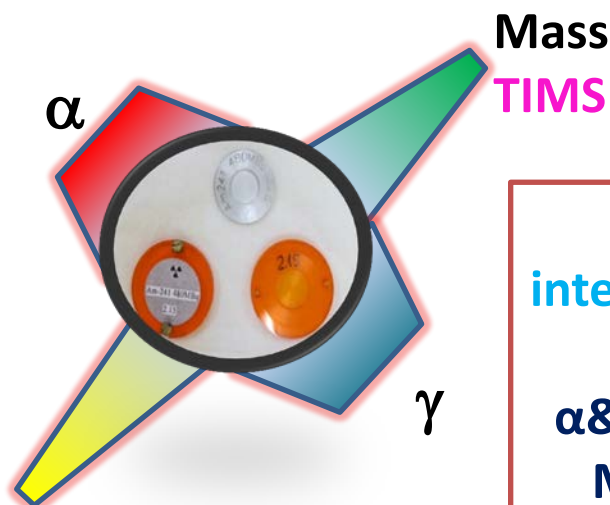
0.1% uncertainty

* Pu correction: small but to be checked

Cross-check, OK
Discrepancy between
Declared and Measured

Details R300 by
Dr. K. Terada

Heat
Micro Calorimetry



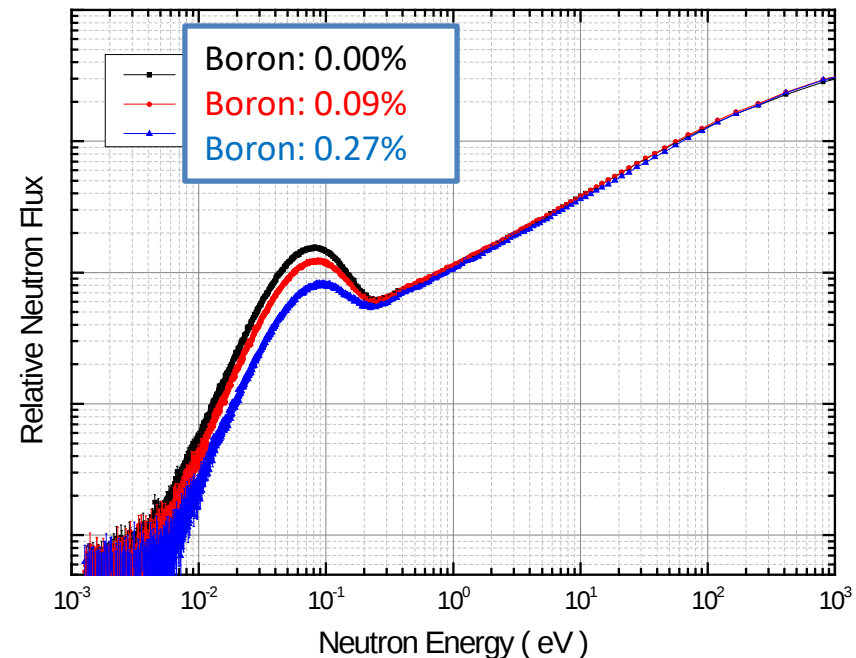
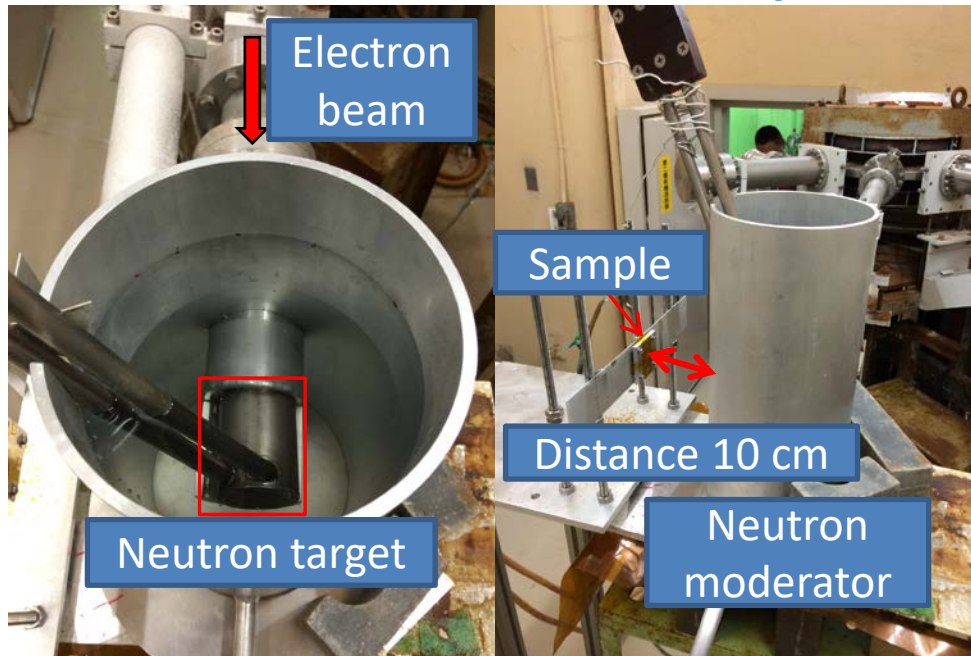
Effectiveness of
integrating independent
methods
 α & γ ray spectroscopy,
Micro Calorimetry,
Mass spectroscopy

② Activation measurement method at Kyoto TOF facility

Development of variable neutron flux field for activation measurements at pulsed neutron facility



KURRI-LINAC



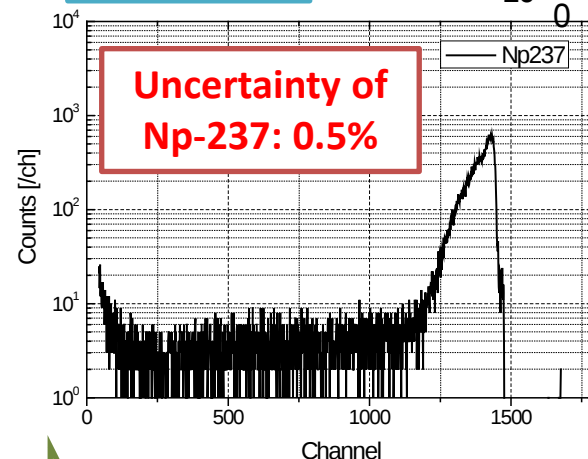
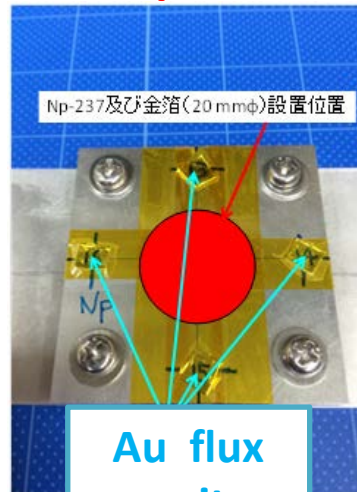
Advantage:

E dependence of neutron flux by TOF method
Variable thermal and epi-thermal flux ratio

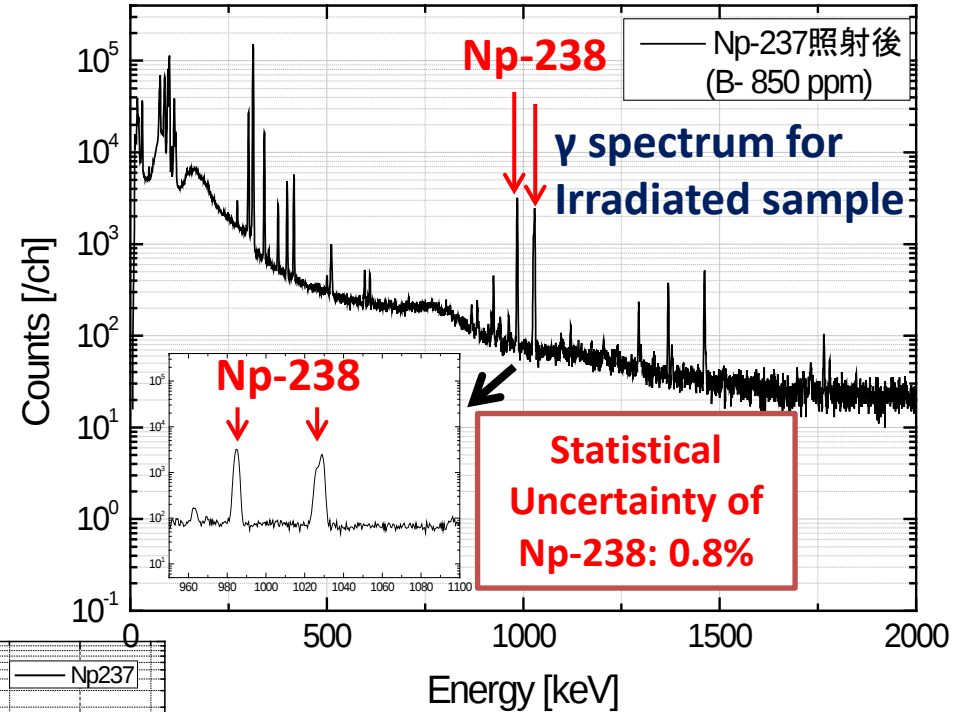
② Activation measurement method at Kyoto TOF facility

Advantage:

Usage of Unsealed Sample



Statistics?



Flux averaged cross section

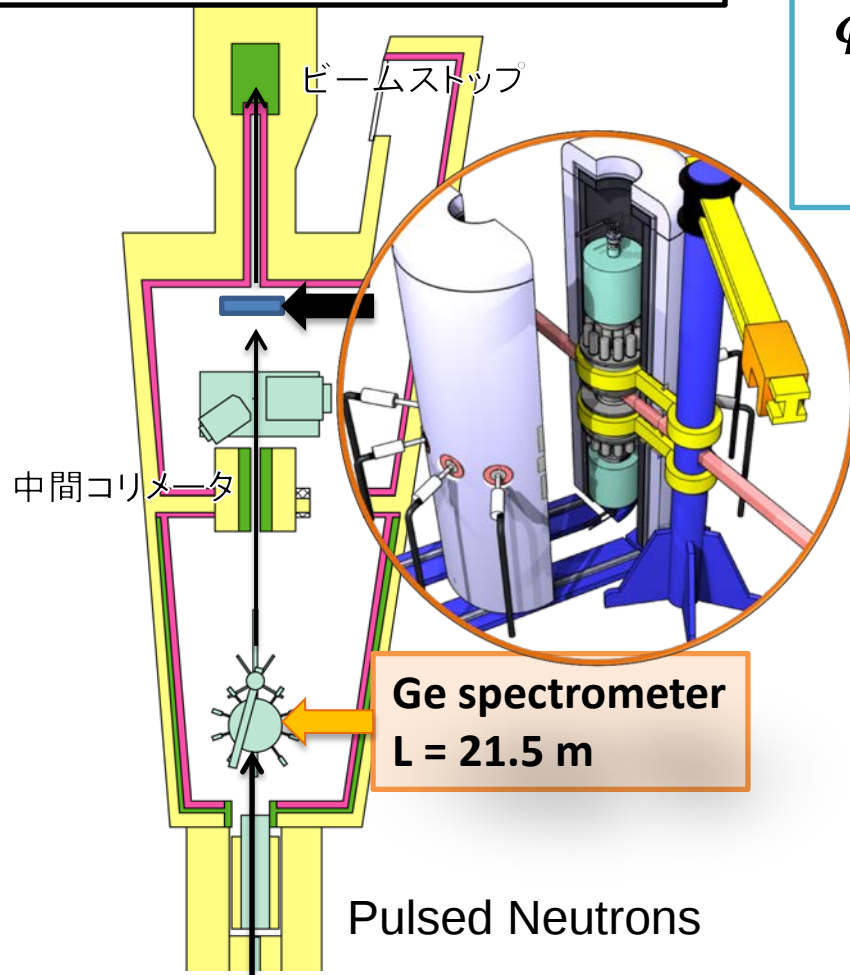
$$\int \phi(E) \sigma_r(E) dE$$

Details by Dr. H. Yashima
(Y. Takahashi) (S033)

Sample amount

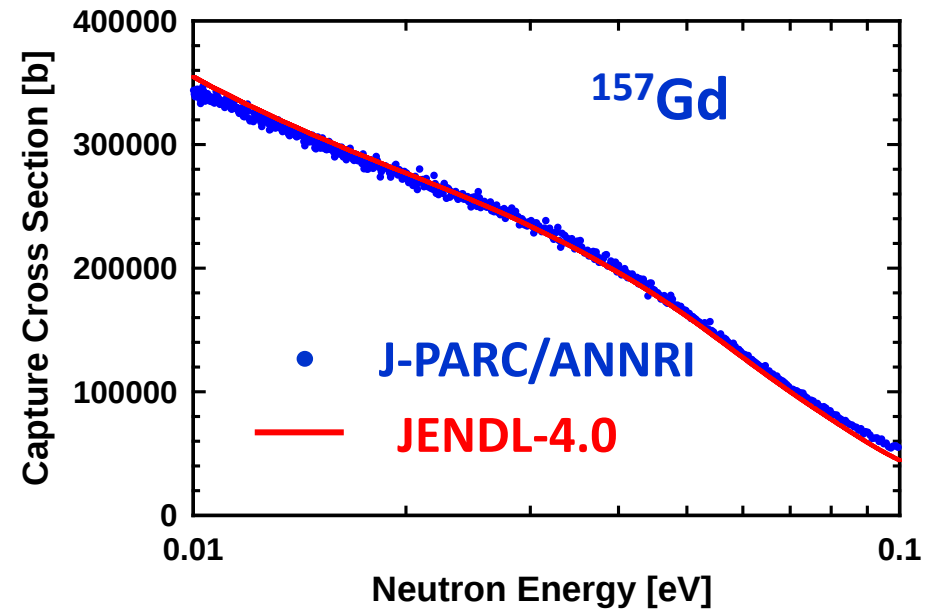
③-1 TOF measurement at J-PARC/ANNRI

A precise absolute value determination method using different thickness samples by Dr. A. Kimura **Gd-155, 157**



Ratio of Capture yields expressed
in the case $\sigma_{\text{cap}} \doteq \sigma_{\text{tot}}$

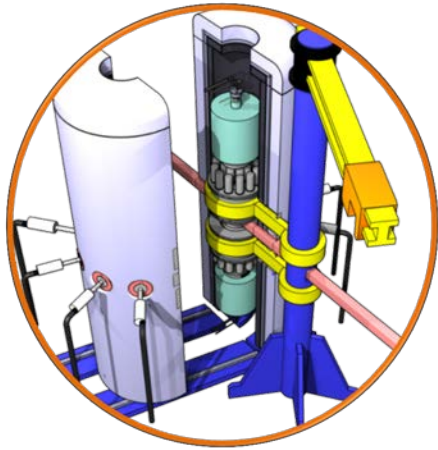
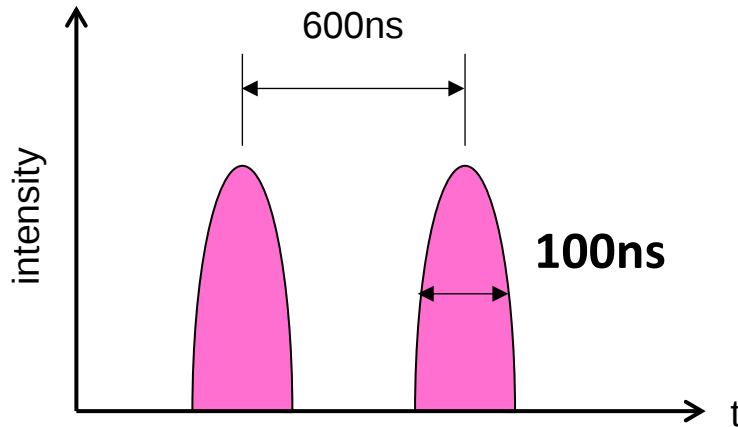
$$\frac{\varphi(E_n) \times k_{Gd} \times (1 - \exp(-\sigma(E_n)N_{\text{thin}}))}{\varphi(E_n) \times k_{Gd} \times (1 - \exp(-\sigma(E_n)N_{\text{thick}}))} = \frac{1 - \exp(-\sigma(E_n)N_{\text{thin}})}{1 - \exp(-\sigma(E_n)N_{\text{thick}})}$$



Details on resonance analyses
by Dr. K. Mizuyama (S114)

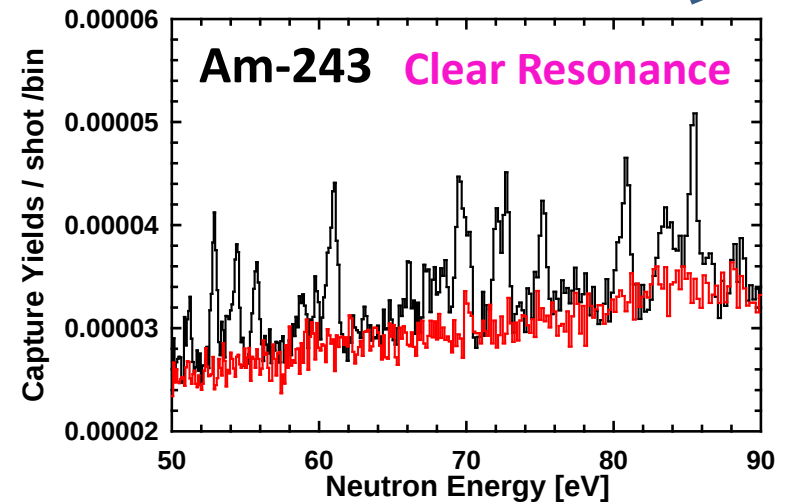
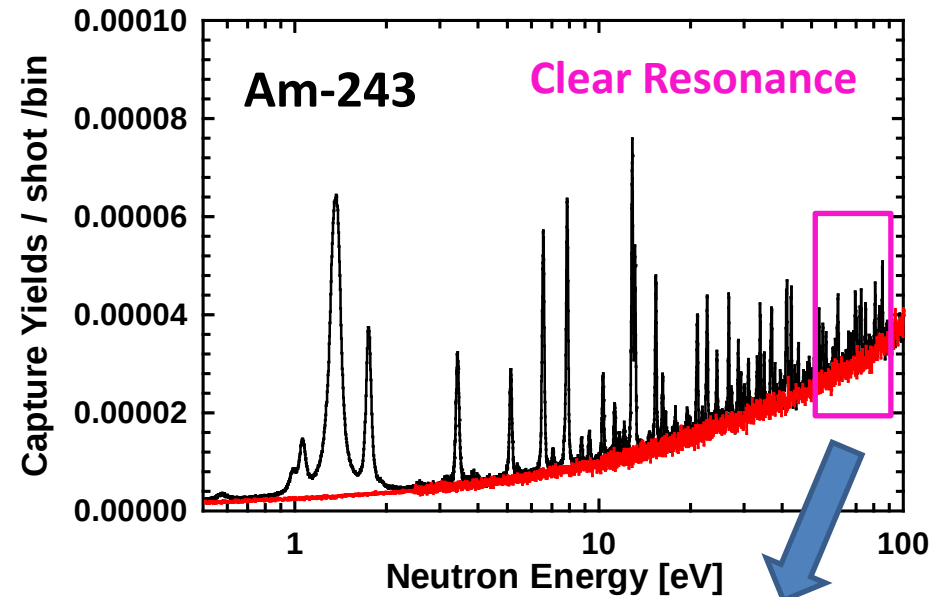
③-2 TOF measurement at J-PARC/ANNRI

J-PARC/ANNRI High Intensity Beam
Double pulse >> **Single pulse**



Ge spectrometer at ANNRI

Single Pulse TOF Data at ANNRI



③-3 TOF measurement at J-PARC/ANNRI

Development of tools for total cross section measurement

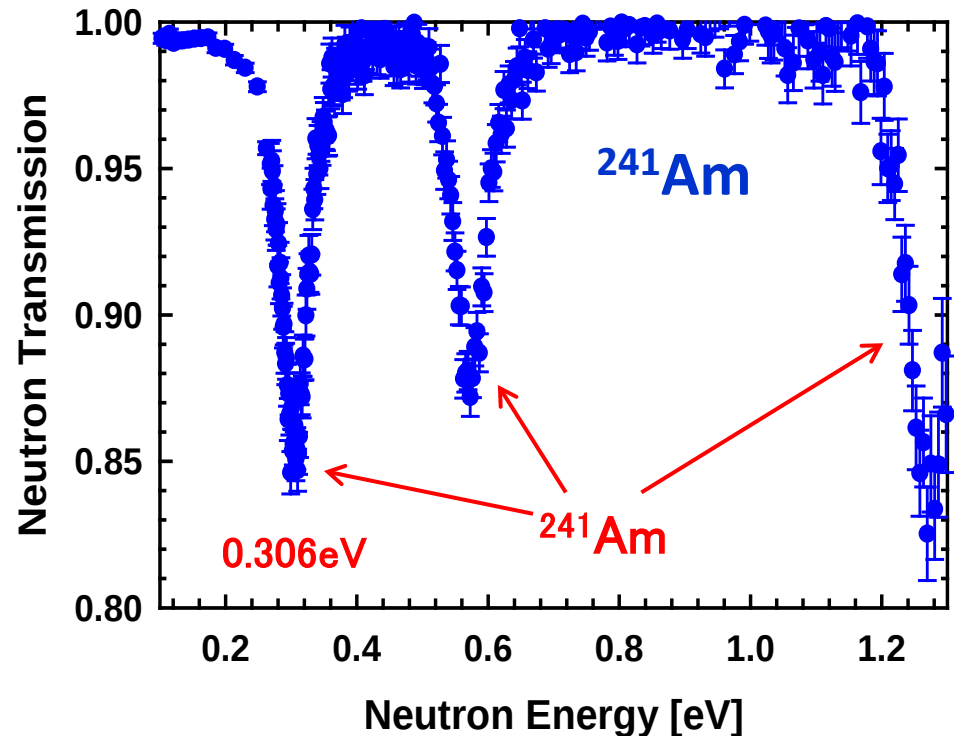
Neutron detectors for Transmission
L = 29 m
(^6Li glass + ^7Li glass)

MA sample

Neutron detector and its DAQ were developed

Details by Dr. T. Nakao (R130)

The first transmission data of MA at ANNRI



Useful info for capture determination
in case Total $\sim$ Capture

③-4 Extension of measurable energy region from 100 keV at J-PARC/ANNRI



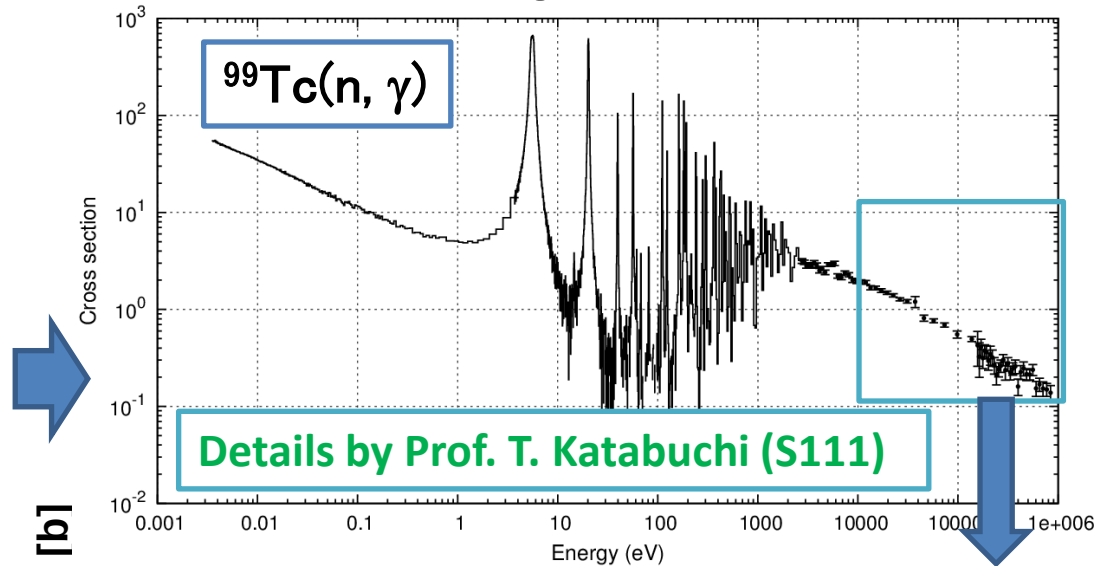
ANNRI-NaI(Tl) spectrometer

DAQ for High counting rate

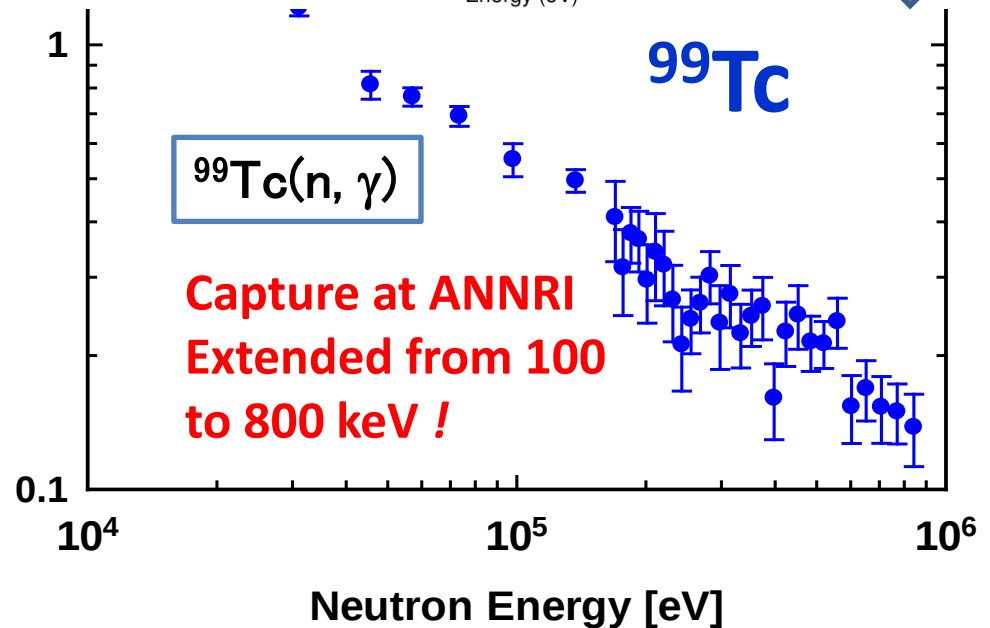
Threshold

Pulse width
measurement
methods

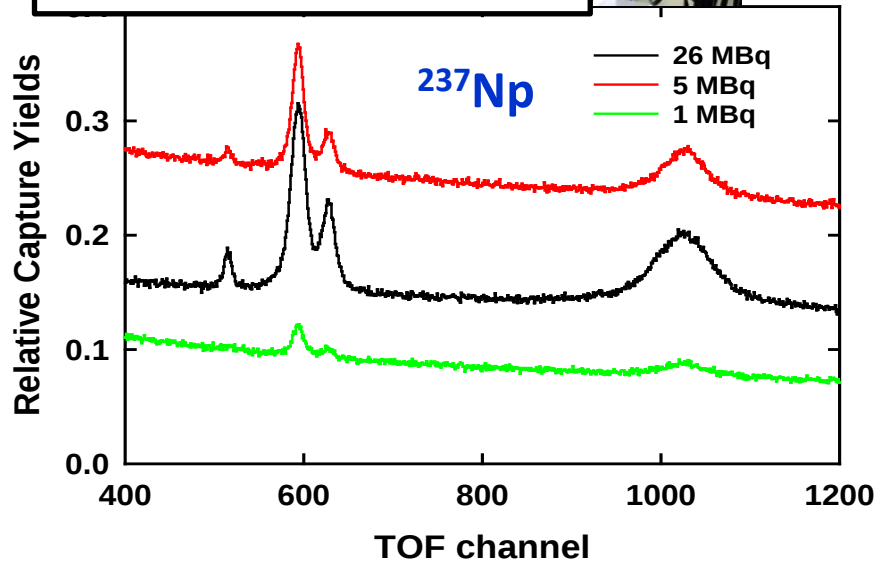
T. Katabuchi, *et al.*,
NIM A764, 369(2014)



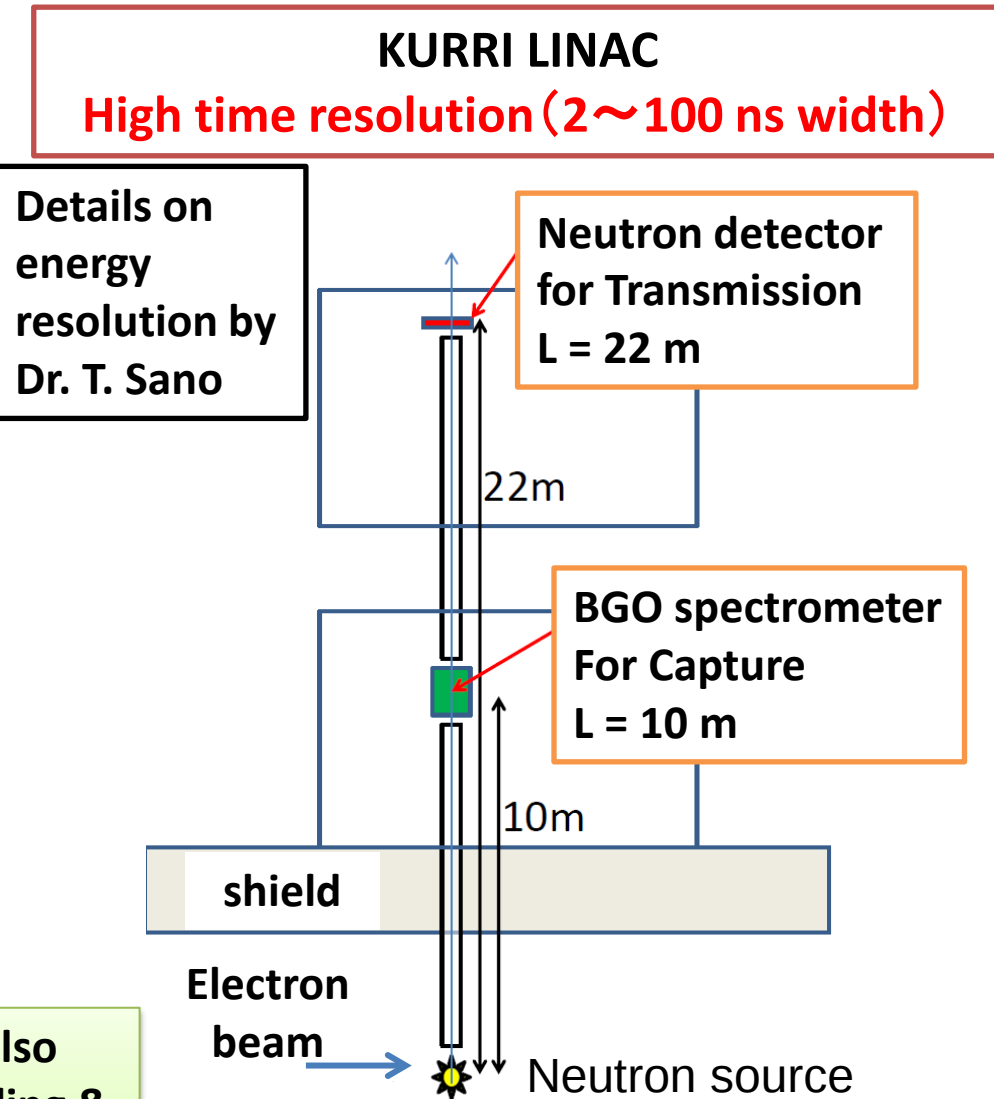
Capture Cross Section [b]



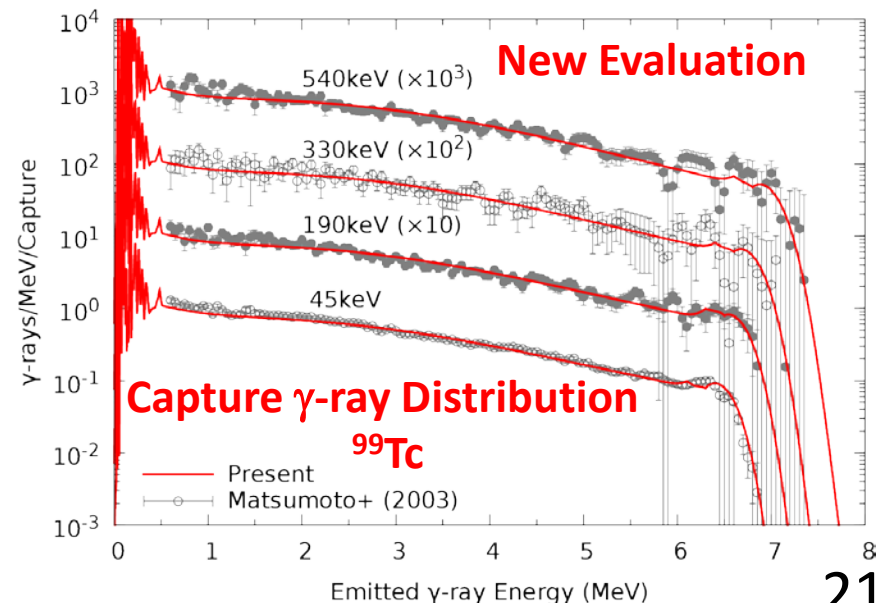
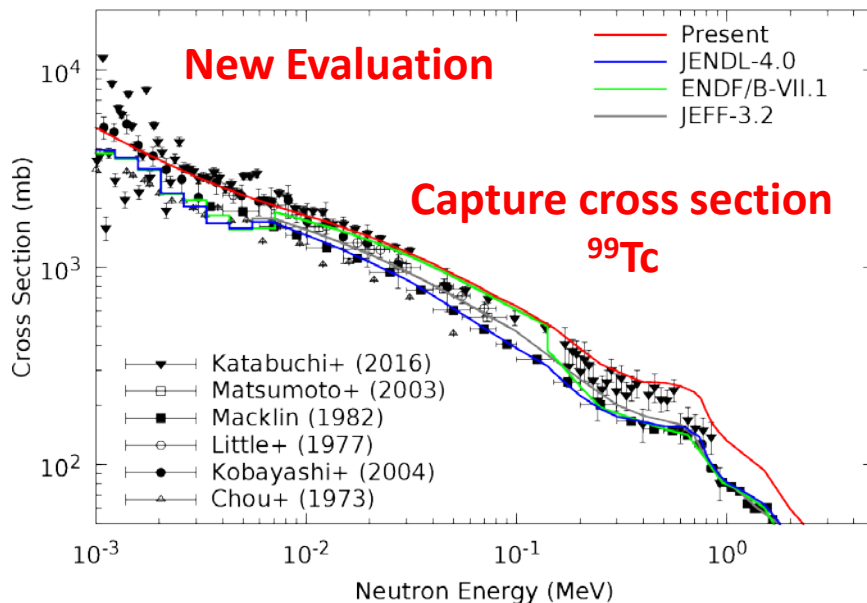
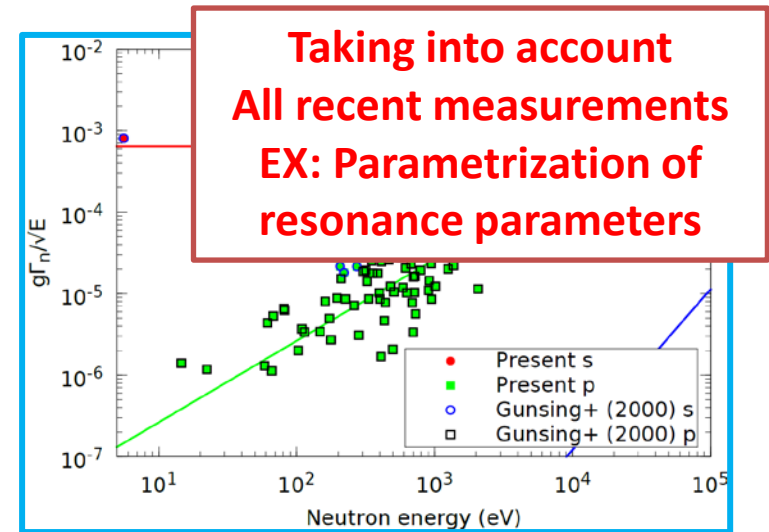
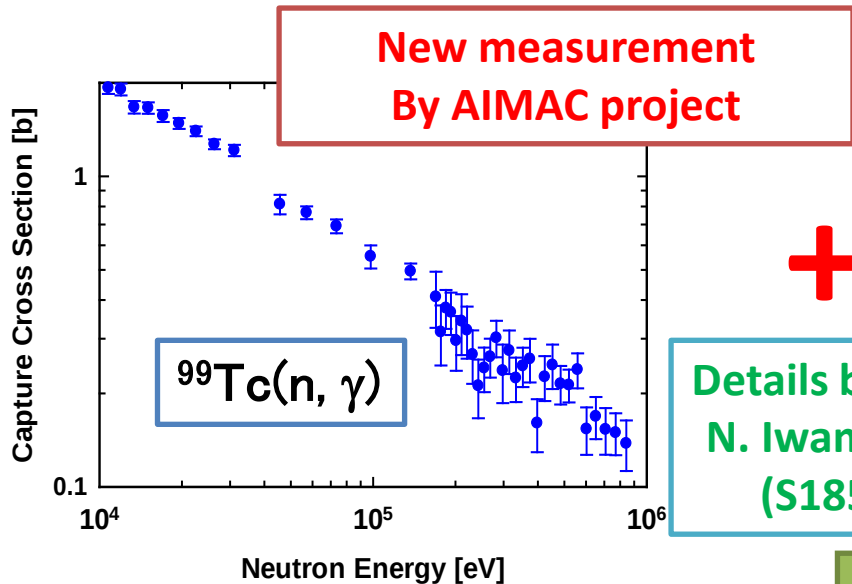
③-5 TOF measurement at KURRI LINAC



Note: Data of different thickness samples also enable systematic cross-check of self-shielding & scattering correction factor



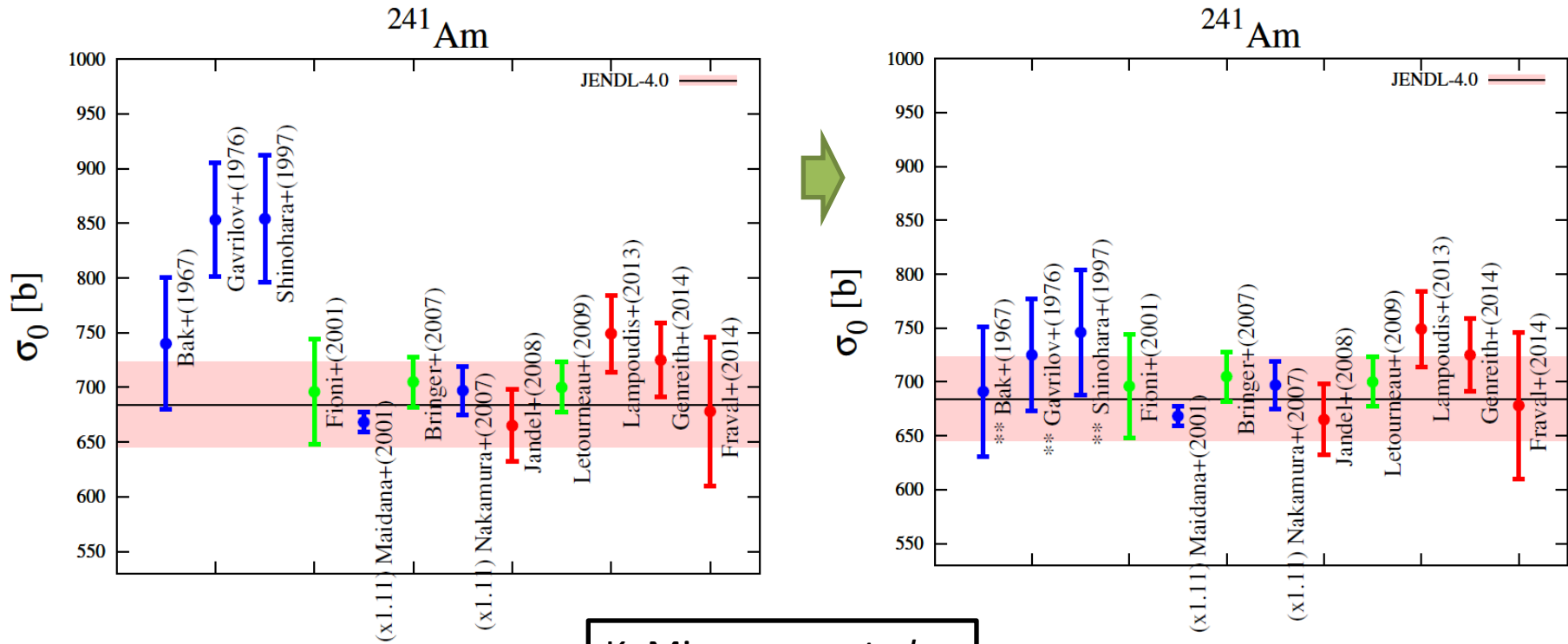
④-1 New Evaluation by AIMAC



④-2 Systematic correction of bias effects

Re-evaluation of Cd ratio (Activation) data

Correction of resonance contribution below Cd cut-off energy



$$\frac{R}{\sigma_0} = g\phi_1 + s_0^c\phi_2 + \delta s_0\phi_2$$

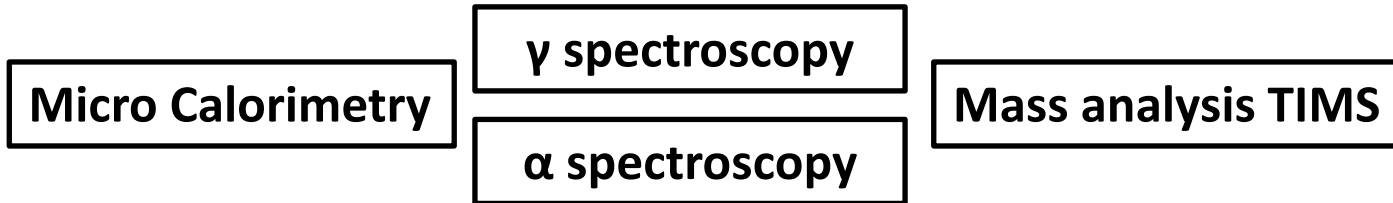
$$\delta s_0 = \frac{2}{\pi^{1/2}\sigma_0} \int_{\mu kT}^{0.5} \frac{dE}{E} \left(\sigma(E) - \sqrt{\frac{E_0}{E}} g\sigma_0 \right)$$

K. Mizuyama, *et al.*,
JNST online (2016)

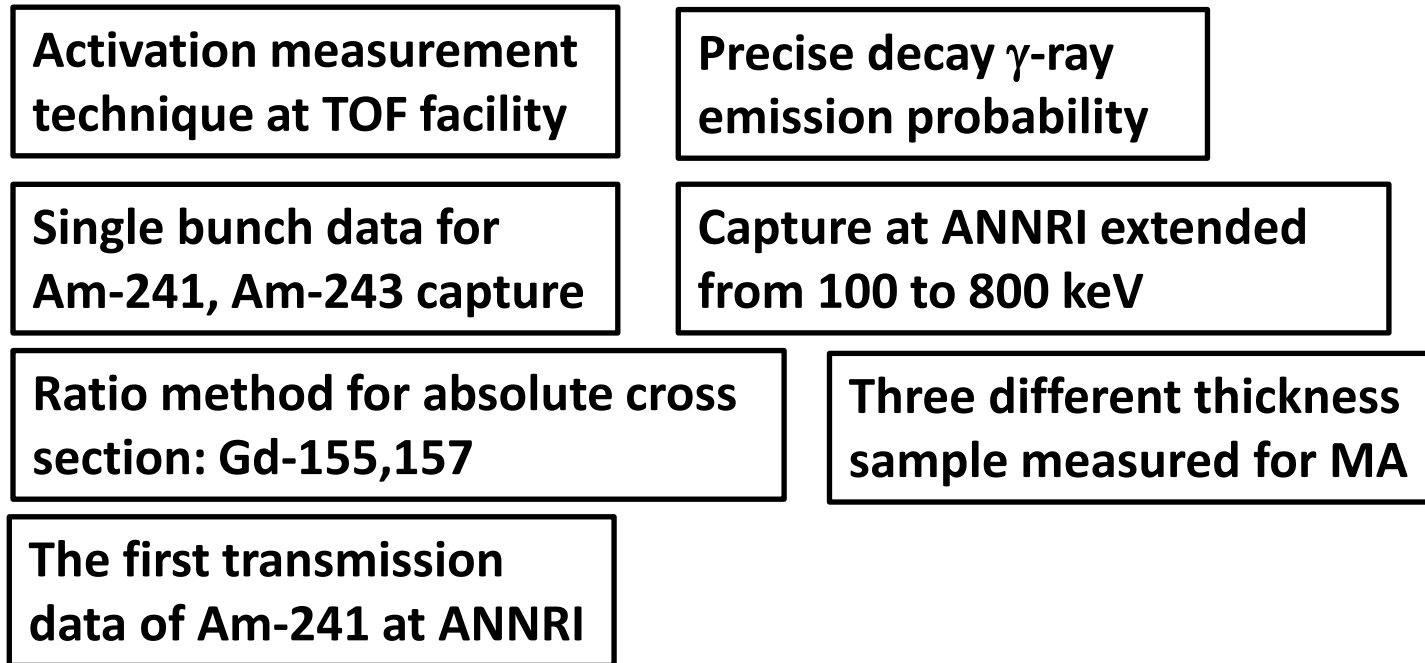
Importance of
identifying bias effects
& systematic correction!

Conclusions

- High precision sample characterization techniques have been developed. Effectiveness of combinational use of these techniques was shown.

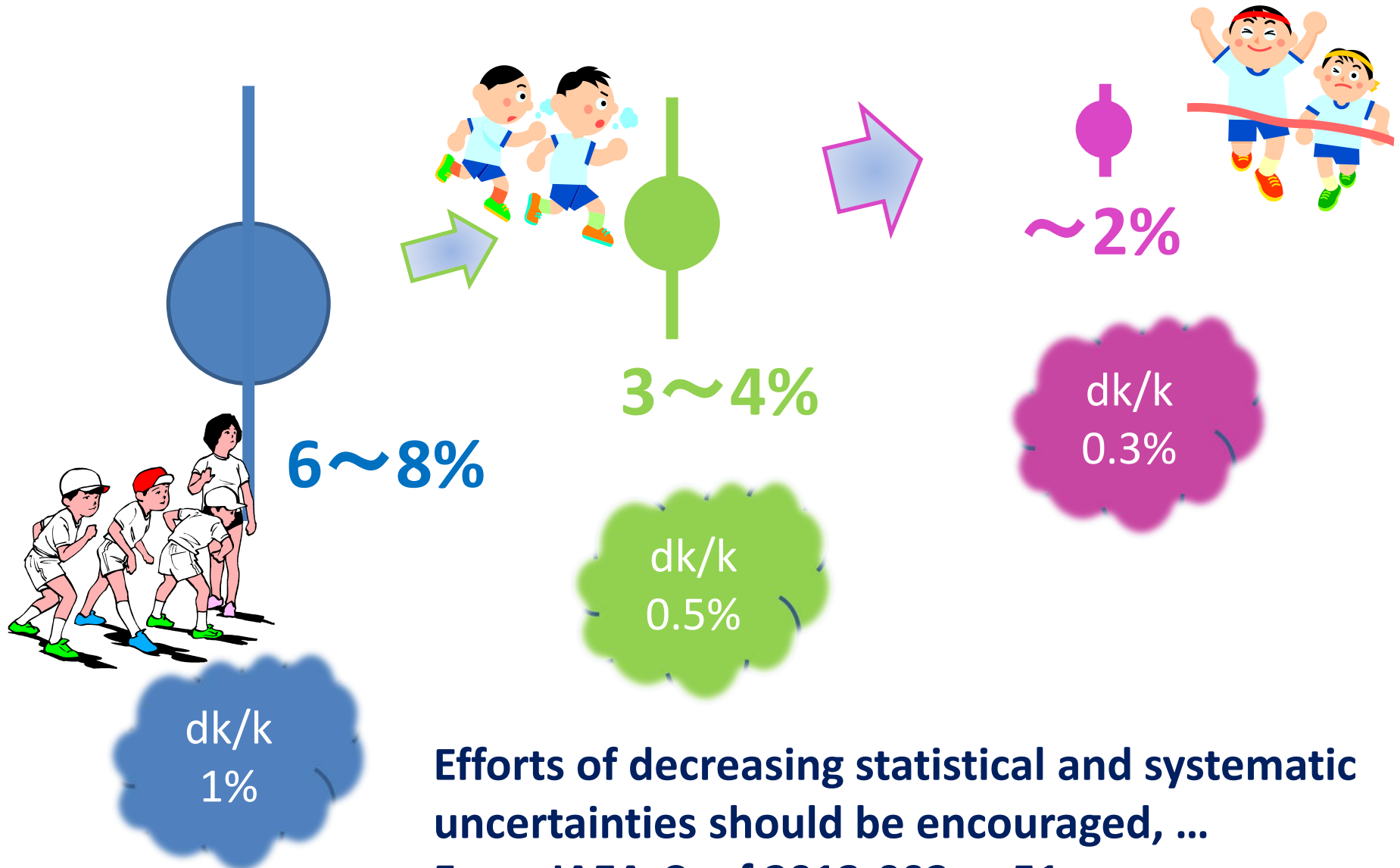


- Highlights of Activation and TOF techniques



- Effectiveness of Re-evaluation with systematic bias effect corrections

Future Perspective



Efforts of decreasing statistical and systematic uncertainties should be encouraged, ...
From JAEA-Conf 2013-002, p.51